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USE OF A STATISTICAL LONG-RANGE TRANSPORT MODEL TO ESTIMATE ONTARIO HYDRO's CONTRIBUTION TO SULFUR AND NITROGEN DEPOSITION

Report No 83-515-K

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ABSTRACT

Ontario Hydro's contribution to wet, dry, and total deposition of SO_2, $SO_4^=$, NO_x and NO_3^- has been estimated using a modified version of a statistical long-range transport model developed by Dr. Bernard Fisher of CEGB. In this report the handling of emissions, deposition, meteorology, and chemistry in the model are described, as well as the model's applicability and limitations. The incorporation of NO_x-NO_3^- chemistry into the model, which was done in this work, is discussed in detail. Model results are compared with measured data.

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To Mr. D.K.A. Gillies
Manager - Environmental Protection
Central Thermal Services

USE OF A STATISTICAL LONG-RANGE TRANSPORT MODEL TO ESTIMATE ONTARIO HYDRO's CONTRIBUTION TO SULFUR AND NITROGEN DEPOSITION

1.0 INTRODUCTION

In 1979, the Atmospheric Research Section acquired a statistical long-range transport model that was developed by Dr. Bernard Fisher of the CEGB (Fisher, 1975 and Fisher, 1978). This model had been written to determine the effects of sulfur emissions from the UK on sulfur deposition over Norway. It was modified for implementation on the Section's HP1000 minicomputer (Dueta, 1981) and for use under North American conditions (McNeil, 1981). Results from a performance and sensitivity analysis (McMillan, 1982) indicated that the model behaviour was generally consistent with the physical processes involved in long-range transport.

Fisher (1975, 1978) included only sulfur emissions, transport and deposition in the statistical transport model. However, Ontario Hydro requires that contributions from nitrogen-containing compounds be included in the estimates of acid deposition. Central Thermal Services (CTS) requested that Fisher's model be modified with the objective of providing an economical in-house method to estimate Ontario Hydro's contribution to sulfur and nitrogen deposition in sensitive areas in and around Ontario. Previously, CTS had been using a regional box model develcped by Acres Consulting Services Ltd, (Acres, 1980).

The following tasks were specified:

(i) obtain emissions inventories for Ontario Hydro and background for 1978-1981

(ii) obtain necessary meteorological data for 1978-1981

(iii) expand the model's grid region to the south and east to include the St. Louis, Kentucky and Boston emissions regions

(iv) incorporate NO_x chemistry

(v) review transformation assumptions

(vi) modify the model output to provide annual wet, dry and total deposition fluxes of SO_2, $SO_4^=$, NO_x, NO_3^-, H^+ and total S and N

(vii) compare the model results with measured data from sampling stations operated by Ontario Hydro

2.0 THE LRT MODEL: BRIEF DESCRIPTION AND SOURCES OF DATA

2.1 Description

2.1.1 Column Density of a Pollutant

The basic quantity calculated in order to evaluate deposition rates is the column density C of a pollutant expected over a particular receptor area which would result from the yearly average emission rate in a particular emitting area, in the absence of removal and transformation processes. This quantity is given by

$$C=fQ/(2\pi xu) \qquad (1)$$

where Q is the emission rate (g y^{-1}), x is the emitter-receptor distance (m), and u is the average wind speed (m s^{-1}). The factor f accounts for the statistical distribution of wind directions; it is equal to the meteorologically observed probability that the wind is blowing from the emitter to the receptor divided by the probability that would result if the wind blew equally often from all directions.

C as computed using equation (1) is expressed in the apparently awkward units of gs $m^{-2}y^{-1}$, but when multiplied by a precipitation scavenging coefficient expressed in s^{-1}, it yields an annual wet deposition flux and when divided by a mixed layer depth (m) and multiplied by a dry deposition velocity (m s^{-1}), it yields an annual dry deposition flux. It is thus a convenient basic quantity to use.

Before C can be used to calculate these fluxes properly it must be corrected to account for the following three removal processes:

(i) dry deposition

(ii) scavenging by precipitation

(iii) chemical transformation

Process (iii) is not strictly a removal process, since $SO_4^=$ and NO_3^- are created when SO_2 and NO_x are removed from the atmosphere by oxidation.

2.1.2 Wet and Dry Deposition Processes

Dry deposition is assumed to occur at the receptor at the rate D(g $m^{-2}y^{-1}$), given by

$$D = Pv_d CG(0,h,x/u)\exp(-kx/u) \qquad (2)$$

where v_d is the dry deposition velocity of the pollutant (m s^{-1}) and C is the column density as given in equation (1). G(0,h,x/u) is a factor (units m^{-1}) which converts the column density to a ground-level (z=0) concentration assuming that the pollutant is released at height h and has travelled a distance x transported by a wind of speed u. G also depends on the mixed layer depth a (m). G is calculated by a twelve-term series approximation (Scriven and Fisher, 1975) to the solution of an atmospheric diffusion equation. A value for the vertical eddy diffusivity K (m^2 s^{-1}) is required to calculate G. G includes the effects of removal along the transport path by dry deposition.

The factor P in equation (2) represents the probability a particle escapes wet deposition along the transport path and arrives at the receptor. Calculation of P is discussed below. The exponential factor in equation (2) represents the fraction of the pollutant which does not undergo chemical transformation; this too is discussed below.

Deposition by precipitation scavenging is modelled as occurring at the receptor at the rate W (g $m^{-2}y^{-1}$), given by

$$W = P\lambda C\{\int_0^a G(z,h,x/u)\,dz\}\exp(-kx/u) \qquad (3)$$

where λ (s^{-1}) is the precipitation scavenging coefficient for the pollutant, and the other quantities are as in equation (2). Equation (3) applies only during precipitation; W is 0 in dry periods. λ is a function of the average precipitation intensity I (mm h^{-1}) during wet periods.

For $SO_4^=$ and NO_3^-, which are for the most part formed in the atmosphere rather than being released at a definite elevation from a stack, the factor CG in equation (2) and the product of C and the integral in equation (3) are replaced by the average concentration in the mixed layer, C/a.

Provision is made in the model for $SO_4^=$ and NO_3^- to be emitted directly, as well as being formed by reaction in the atmosphere. This is parameterized by assuming that a certain fraction F of the emitted SO_2 or NO_x is emitted as $SO_4^=$ or NO_3^-.

2.1.3 Probability of Washout Along the Transport Path

The probability P that a particle survives wet deposition to reach the receptor region is calculated using a Markovian model (Fisher, 1975, 1978) in which the parameters are the probabilities per unit time of a transition from a dry to a wet period and from a wet to a dry period. The transition probabilities are the reciprocals of the average durations t_0 and t_1 of dry and wet periods, which can be determined from hourly precipitation data. P is also a function of λ, the precipitation scavenging coefficient.

2.1.4 Chemical Transformation

Chemical transformation is assumed to occur in the gas phase at a rate proportional to concentration of the species being consumed. SO_2 and NO_x are transformed to $SO_4^=$ and NO_3^- by reaction. These are parameterized by first-order rate constants k (s^{-1}). If the species whose dry or wet deposition is studied is formed rather than consumed by the reaction, the exponential factors in equations (2) and (3) are replaced by somewhat more complicated expressions.

2.1.5 Summary

The meteorological parameters u, a, and K vary with the seasons; these variations are accounted for in the model by calculating D and W as weighted sums over seasonal deposition rates.

With the transport, removal and deposition processes modelled as described above, the parameters and data necessary to run the model are the meteorological parameters f, u, a, K, t_0, t_1, and I; the emission rates Q_{SO_2} and Q_{NO_x} throughout the region and the emission parameters h, F_{SO_4} and F_{NO_3}; the deposition parameters v_d and λ for SO_2, $SO_4^=$, NO_x and NO_3^-; and finally the chemical rate constants k_{SO_2} and k_{NO_x}.

The remainder of this section describes the sources for the meteorological and emissions data; the chemical and deposition parameters are discussed in the succeeding section.

2.2 Emissions

The Fisher statistical long-range transport model was developed in order to determine the annual (long-term) SO_2 deposition fluxes over Norway (Fisher, 1975, 1978). The emission region considered by the model was a 22 x 22 grid over Europe, with grid squares 127 km on each side. The annual SO_2 emission rate for each grid square was set equal to the sum of annual emissions from all sources within the square and was taken to be emitted from the centre of the square.

In modifying the model for use under North American conditions (McNeil, 1981), the grid was changed to an 18 x 18 grid which covered the northeastern United States and southeastern Canada with 100 km grid squares (Figure 1). In this work the model's grid region was expanded to the south and east to form a 22 x 22 emission grid (Figure 2) with the squares still 100 km on each side. In addition to SO_2 emission rates in this emission area expressed as kilotonnes SO_2 emitted within each square per year, the NO_x emissions in each square are now included as the equivalent mass in kilotonnes of NO_2 emitted per year.

The emission rates used for both SO_2 and NO_x for sources other than fossil fuel-fired generating stations operated by Ontario Hydro are identical to those used by Acres Consulting Ltd in runs of their Regional Box Model for CTS (1978 emissions from runs described in Acres, 1980; other years from raw output supplied by CTS).

Ontario Hydro's contribution to sulfur and nitrogen oxide emissions were calculated using annual emission rates from generating stations which were operated during the years 1978-1981 (Reeves, 1979; Cross, 1980; Reeves, 1981; Perks, 1982). Ontario Hydro's emissions of nitrogen oxides, given as mass of NO emitted in the Ontario Hydro reports, were converted to mass of NO_2 for input to the model.

Initially, the incorporation of seasonal values of emissions was considered important. However, upon investigation, it appears that emissions from most significant sources are approximately equal for the four seasons (Jarv and Surtees, 1982). Consequently, it was decided to use one emissions inventory per year.

A specified fraction of emissions are assumed to occur at ground level (z=0) while the remainder are assumed to occur at a height determined by wind speed. Assumed release heights for emissions are summarized in Table I. These values were taken without modification from Fisher (1975).

TABLE I

ASSUMED RELEASE HEIGHTS FOR ALL EMISSIONS

Fraction of low level sources:	0.5
Effective stack height (m) of high-level sources:	$h = 200 + \frac{1000}{u}$,

u is average wind velocity, m s^{-1}

It was assumed initially, following Fisher (1975), that 10% of the emissions nominally as SO_2 were actually emitted as $SO_4^=$; it was assumed that none of the NO_x emissions were in the form of NO_3^-.

2.3 Meteorology

The statistical model uses long-term statistical distributions of wind speed, wind direction, dispersion categories, and rainfall to determine the annual wet and dry deposition fluxes of sulfur and nitrogen. The main assumption behind the model is that details of the fluctuations of meteorological and physical conditions within a season have only a small influence upon the overall average deposition fluxes. Therefore, the model is an efficient way of obtaining average distribution patterns. Sensitivity studies have shown that greater emphasis on the smaller-scale fluctuations would have little effect on annually averaged results (McMillan, 1982).

2.3.1 Mixing Parameter Sets

The model requires sets of meteorological input data consisting of an average wind speed, mixing height, eddy diffusivity, and a weighting factor which describes the fraction of the year during which that meteorological condition (defined in Table II) is expected to occur.

TABLE II

MIXING PARAMETER SETS

Season	Stability Class	Meteorological Input Data			
		Wind Speed (m/s)	Mixing Height (m)	Diffusivity (m^2/s)	Probability of Occurrence Within the Year
Summer	Stable	6.7	450	3	0.167
	Unstable	5.65	1 300	30	0.083
Fall	Stable	7.8	468	4	0.1875
	Unstable	4.6	1 000	30	0.0625
Winter	Stable	7.38	450	4	0.21875
	Unstable	4.34	900	30	0.03125
Spring	Stable	8.3	500	4	0.125
	Unstable	5.7	1 100	30	0.125

The dispersion of pollutants within the mixed layer is determined in the model as a function of the wind speed, mixing height, and eddy diffusivity. Estimates of the mixing height and eddy diffusivity are based on stability class and wind speed. The stability is classified as being either mechanical (stable) or convective (unstable). Eight data sets were chosen for this study to describe unstable and stable conditions for each season. The derivation of these data, which are presented in Table II, is described in detail in McMillan (1982).

2.3.2 Wind Direction

Fisher's model used a weighting factor assigned to each of 8 equal wind sectors to take wind direction into account. In this work, values for ground-level wind frequency distribution for 16 equal wind sectors were taken from the Canadian Normals wind report for 1955-1972 (Environment Canada, 1975; McMillan, 1982). The wind rose constructed from this frequency distribution is shown in Figure 3. The frequency distribution itself is presented in Table III. The wind data presented here are taken to characterize the whole gridded region shown in Figure 2.

2.3.3 Rainfall

Values for t_ϕ and t_γ, the mean lengths of wet and dry periods, were taken from Venkatram et al, 1982. The inverses of these values are summarized in Table IV, along with the assumed average precipitation intensity during wet periods. These values were assumed to apply to the entire gridded region.

TABLE III

FREQUENCY DISTRIBUTION AND SECTOR ANGLE DEFINING WIND ROSE

Sector Number	Angle (radians)	Weighting Factor Based on Probability of Wind in Sector
1	-2.945 to -2.552	1.24
2	-2.552 to -2.160	1.42
3	-2.160 to -1.767	1.00
4	-1.767 to -1.374	1.16
5	-1.374 to -0.982	0.62
6	-0.982 to -0.589	0.67
7	-0.589 to -0.196	0.54
8	-0.196 to 0.196	0.89
9	0.196 to 0.589	0.64
10	0.589 to 0.982	0.77
11	0.982 to 1.374	0.64
12	1.374 to 1.767	1.01
13	1.767 to 2.160	0.90
14	2.160 to 2.552	1.42
15	2.552 to 2.945	1.36
16	2.945 to -2.945	1.75

TABLE IV

SUMMARY OF PRECIPITATION DATA FOR FISHER's MODEL

Inverse of mean length of dry period:	0.60×10^{-5} (s^{-1})
Inverse of mean length of wet period:	0.40×10^{-4} (s^{-1})
Assumed rainfall intensity:	1 mm h^{-1}

2.3.4 Summary

All of the above meteorological data were derived from observations over several years. No attempt has been made to extract meteorological parameters specific to a given year in this work.

2.4 Chemical Parameters

The chemical parameters which must be specified to calculate average deposition fluxes of sulfur and nitrogen compounds are the dry deposition velocities for SO_2, $SO_4^=$, NO_x, and NO_3^-, the precipitation scavenging coefficients for the same four species, and two first-order rate coefficients describing the transformation of SO_2 to $SO_4^=$ and of NO_x to NO_3^-.

2.4.1 Precipitation Scavenging

As Fisher (1978) has pointed out, within the Markovian procedure used to calculate the extent of wet removal during transport in this model, it is necessary to assign the same precipitation scavenging coefficient to interconvertible compounds (such as SO_2 and $SO_4^=$ or NO_x and NO_3^-); otherwise, deposition probabilities would depend upon where along the transport path wet or dry periods occur. This information is not available within the stochastic framework adopted in Fisher's model, and therefore the calculation of loss through wet deposition is valid only if the rate of wet deposition is independent of the degree to which chemical transformation has occurred.

For SO_2 and $SO_4^=$ this is not a serious restriction, because although the $SO_4^=$ scavenging coefficient exceeds that for SO_2 by about a factor of 10, a higher assumed scavenging coefficient for SO_2 can compensate for in-cloud oxidation. For NO_x and NO_3^- the restriction is severe, because NO and NO_2 are practically insoluble in water (Schwartz and White, 1981), and are probably not oxidized to an appreciable extent in-cloud (Lee and Schwartz, 1981).

It was decided to use a single effective scavenging coefficient for NO_x and NO_3^- rather than to go outside the probabilistic scheme upon which the statistical LRT calculation is based.

The gases SO_2, NO, and NO_2 are only sparingly soluble in water and are thus scavenged by precipitation in equilibrium rather than irreversibly (Hales, 1972). Their washout kinetics are more appropriately parameterized with a washout ratio than with a scavenging coefficient. However, the scavenging coefficient parameterization, which is the correct one for $SO_4^=$ and for NO_3^-, was used for all compounds in this work.

2.4.2 Sulfur Parameters

The values initially adopted for the transformation and deposition parameters for SO_2 and $SO_4^=$ are presented in Table V.

The values for the dry deposition velocities adopted in Table V are lower than the experimentally measured values for SO_2 and for sulfate aerosol by about 50%. This reduction is made to account roughly for the isolation of much of the transport layer from the surface by a thermal inversion during the night (Fisher, 1978).

The values given for the scavenging coefficients are those that apply for a precipitation intensity of 1 mm h^{-1}, which was assumed over the entire gridded region in this work. For this work the single scavenging coefficient for SO_2 and $SO_4^=$ was taken to be equal to the measured scavenging coefficient for sulfate aerosol, the higher of the two scavenging coefficients, as was done by Fisher (1978). The justification for picking the higher value, rather than an intermediate value, is that this choice of an unrealistically high scavenging coefficient for SO_2 accounts roughly for the accelerated oxidation of SO_2 which takes place in the aqueous phase in clouds. The effect of the oxidation is to convert SO_2 to the more rapidly scavenged $SO_4^=$; attempts to account for the accelerated oxidation by increasing the transformation rate constant tend to work less well because the rate coefficient applies in the model during both wet and dry periods, while in-cloud oxidation occurs to a much greater extent in wet periods (Venkatram et al, 1982).

2.4.3 Nitrogen Parameters

Values of deposition parameters for nitrogen compounds are in general known less precisely than are those for sulfur compounds; values given in the literature for the necessary parameters for NO_x and NO_3^- are summarized in Table V.

The values given for the dry deposition velocities in Table V are values recommended for modelling removal of NO_x and NO_3^- from a dispersing plume (Bottenheim and Strausz, 1982); as such they probably include to some extent the reduction discussed in the previous subsection made to account for the nighttime isolation of the transport layer from the earth's surface. Indeed, some recently reported experimental values for the dry deposition velocities of NO_3^- (0.2-1.0 cm s^{-1}, Hicks et al, 1983) and HNO_3 (2.5 ± 0.9, Huebert, 1983) exceed the values given in Table V.

TABLE V

INITIALLY CHOSEN CHEMICAL PARAMETERS

SULFUR PARAMETERS

λ_{SO_2} = $\sim 10^{-5}$ s^{-1} (Garland et al, 1978)

λ_{SO_4} = $\sim 10^{-4}$ s^{-1} (Garland et al, 1978)

v_d (SO_2) = 0.5 cm s^{-1} (Fisher, 1978)

v_d $(SO_4^{=})$ = 0.05 cm s^{-1} (Venkatram et al, 1982)

k $(SO_2 \rightarrow SO_4^{=})$ = 0.01 h^{-1} (Fisher, 1978)

Used for base runs: as above, but

v_d $(SO_4^{=})$ = 0

$\lambda_{SO_2} = \lambda_{SO_4} = 10^{-4}$ s^{-1}

NITROGEN PARAMETERS

λ_{NO_x} = 2.5×10^{-6} s^{-1} (Bottenheim and Strausz, 1982)

$\lambda_{NO_3^-}$ = 5×10^{-5} s^{-1} (Bottenheim and Strausz, 1982)

v_d (NO_x) = 0.2 cm s^{-1} (Bottenheim and Strausz, 1982)

v_d (NO_3^-) = 1.0 cm s^{-1} (Bottenheim and Strausz, 1982)

k $(NO_x \rightarrow NO_3^-)$ = 0.1 h^{-1} (Bottenheim and Strausz, 1982)

Used for base runs: as above, but

$\lambda_{NO_x} = \lambda_{NO_3}$ = 5×10^{-5} s^{-1}

v_d (NO_3^-) = 1.0 cm s^{-1}

Nitrate aerosol is scavenged by precipitation at roughly half the rate of sulfate aerosol and NO_x at roughly one-quarter the rate of SO_2 (Bottenheim and Strausz, 1982). The NO_3^- scavenging coefficient, the higher of the two, was chosen to represent precipitation scavenging of both NO_x and NO_3^-, in order that nitrate aerosol concentration at a given site might correlate well with nitrate wet deposition fluxes at the same site. However, the argument that an unrealistically high scavenging coefficient for NO_x compensates for accelerated oxidation in cloud cannot be applied because NO_x is probably oxidized only to an extremely small extent in the aqueous phase (Lee and Schwartz, 1981). This problem is discussed more fully in Section 5.

3.0 INITIALLY-CHOSEN CHEMICAL PARAMETERS: CALCULATIONS AND RESULTS

Calculations of the total annual wet deposition fluxes of SO_2, $SO_4^=$, NO_x, and NO_3^- for 1979 were performed for receptor squares where Ontario Hydro monitoring stations were located; ambient air concentrations of $SO_4^=$ and NO_3^- were also calculated. The year 1979 was chosen because of the availability of both an accurate emissions inventory and relatively widespread monitored data for that year (Jarv, 1982). The chemical parameters used for these calculations were those given in Table V. The results for wet deposition are given in Table A.1 and for ambient air concentrations in Table A.2, in Appendix A. Also shown are the values observed at the Ontario Hydro monitoring stations (Jarv, 1982). In calculating the modelled wet deposition of $SO_4^=$ it was decided to sum the wet deposition fluxes of SO_2 and $SO_4^=$, since in the precipitation analyses no attempt was made to prevent oxidation of SO_2 in the collected precipitation. No comparison of modelled dry deposition fluxes is done because of the unavailability of dry deposition flux measurements.

The modelled values in Table A.1 for sulfate wet deposition agree rather weakly with the observed values, with the modelled values typically lower than the observed values by about 40-50%. For ambient air concentrations (Table A.2),the agreement is somewhat better, with the modelled values typically higher than the observed values by about 30%.

For nitrogen-containing compounds the agreement is much worse. The nitrate wet deposition is typically much lower than the observed values; the modelled values being approximately 45% of the observed values (Table A.1) (wet deposition of NO_x is essentially zero because of the low solubility of these gases). For ambient air concentrations the modelled values are much larger than the observed values, often by factors of 8-13.

Two monitoring sites, Abitibi and Essex, showed somewhat different patterns in the comparisons between modelled and observed values, as follows:

(1) Modelled $SO_4^=$ wet deposition (Table A.1) is underestimated more severely at Abitibi (grid square 10,17) than at other sites. Small local sources not included in the emissions inventory may account for a significant fraction of the measured $SO_4^=$ wet deposition and air concentration at this remote site. This would explain the extreme underestimation of wet deposition of $SO_4^=$ at Abitibi when compared to the results for the other locations. Modelled and monitored values of NO_3^- wet deposition and air concentration at Abitibi do not show a similar effect.

(2) In Table A.2, the calculated $SO_4^=$ and NO_3^- air concentrations at the Essex site (grid square 9,8) are overestimated to a lesser degree than at the other locations. This may be due to the location of this site within the urban plume of Detroit-Windsor. It has been reported that measurements at Essex are particularly difficult to reproduce using long-range transport models (Olson and Oikawa, 1983).

4.0 INITIALLY-CHOSEN CHEMICAL PARAMETERS: DISCUSSION

Fisher (1983) proposed that prediction of deposition fluxes to within a factor of two of the observed values is the closest agreement that can be expected of simple statistical models. However, the agreement of the modelled wet deposition fluxes with observed values in Tables A.1 and A.2 is not sufficiently good that the values could be used directly for meaningful estimates of acid deposition. Although wet deposition fluxes tend systematically to be underestimated at the sites available for comparison, the degree of underestimation is not consistent enough that multiplication of the modelled values by a single correction factor would give a meaningful estimate.

Fisher's model when applied to Europe encountered similar problems. A recent assessment of sulfur deposition over Ireland showed modelled wet deposition fluxes considerably lower than the observed values (Fisher, 1982). This discrepancy was attributed to wet deposition of natural sulfur (excluding sea-salt sulfate, which was corrected for in the observed values), to sulfate from fertilizers, and to transatlantic transport of anthropogenic sulfur from Canada and the USA. The possibility of fundamental inaccuracies in the model was not discussed by Fisher (1982).

The North American results of Tables A.1 and A.2 cannot be explained in terms of a background of emissions omitted from the inventory because background emissions would tend to cause both wet deposition and ambient air concentrations of sulfate to be underestimated by the model, whereas in fact the model underestimates wet deposition and overestimates ambient air concentrations.

Thus, the model apparently represents one or more aspects of transport, transformation, or deposition incorrectly. The reasons for this are not apparent and will require careful consideration to discover. However, several observations can be made:

(1) Since ambient air concentrations are overestimated and wet deposition underestimated at the same sites, either the scavenging coefficients or the assumed mixed layer depths are too small.

(2) Unrealistically small values for the scavenging coefficients would result from an underestimated average precipitation intensity during wet periods. However, meteorological data tend to confirm the value 1 mm h^{-1} (Venkatram et al, 1982).

(3) High modelled ambient air concentrations of sulfate and nitrate at sites relatively close to industrial emitters can result from assumption of too large a fraction emitted in the oxidized form or from overestimates of the oxidation rates.

(4) High modelled airborne concentrations can also result from underestimated dry deposition rates for the unoxidized form.

(5) Assumption of too high a release height would tend to cause underestimation of dry deposition rates for the unoxidized form.

(6) Since the NO_3^- scavenging coefficient is used for both NO_x and NO_3^- over the entire transport path, it is expected that NO_x wet deposition rates are overestimated. This would lead to underestimated airborne nitrate concentrations and overestimated nitrate wet deposition. The latter was observed but the former was not.

5.0 ALTERED CHEMICAL PARAMETERS: CALCULATIONS AND RESULTS

5.1 Introduction

Without doing detailed computational tests of the handling of all of the modelled transport, transformation and deposition processes, it is of questionable value to experiment with altered values of parameters. It is uncertain what defect in the representation of the physical processes one is compensating for by changing the value of a given parameter.

Nonetheless, it was decided to attempt several such changes for the following reasons:

(a) If a good fit to observed values is obtained with parameters differing from the base runs (Tables A.1 and A.2) by less than the uncertainties in the experimental values, then the altered set is no less valid than the initial set. However, the model would have to be viewed with caution if such a large sensitivity were observed to the value of a parameter not known with precision.

(b) The assignment of the NO_3^- precipitation scavenging coefficient to NOx is obviously unrealistic. Altering the treatment of NO_3^--NO_x scavenging in some way would not lessen the physical validity of the model.

(c) Because the objective of this work is to produce a model suitable for estimating Ontario Hydro's contribution to total acid deposition, a model which can reliably predict S and N deposition fluxes at Ontario sites is of value to Ontario Hydro, even if the physical basis for the accurate prediction is not apparent.

5.2 Sulfur Parameter Adjustments

The $SO_4^=$ dry deposition velocity was left at zero because the current version of the program does not input a value for this parameter. This could be easily changed. The parameters varied were the SO_2-$SO_4^=$ precipitation scavenging coefficient, the SO_2 dry deposition velocity, the SO_2 oxidation rate constant and the fraction emitted as $SO_4^=$.

5.3 Nitrogen Parameter Adjustments

Nitrogen parameters adjusted were the NO_X and NO_3^- dry deposition velocities, the NO_X-NO_3^- scavenging coefficient, and the NO_X oxidation rate constant. The fraction emitted as NO_3^- was left at 0.

To resolve the problem of the unrealistic modelling of NO_X precipitation scavenging, it was decided to introduce a reduced scavenging coefficient along the transport path, but to retain the nitrate value to describe washout at the receptor site. This allows reduced washout in the early stages of transport when the emissions are principally as NO_X and a higher rate at the end of the transport path when the mixture is principally NO_3^-. This is not a rigorously correct way to model washout of the mixture, since a receptor square close to an emitter square will have its washout calculated using a higher scavenging coefficient than would a location near the end of a long transport path. Nonetheless, this approach provides a way to accommodate the differing washout rates of NO_3^- and NO_X without assigning different coefficient values for the different chemical species.

One problem which arises in this treatment of NO_X-NO_3^- scavenging is the following. For two receptor squares which lie at different distances along the same transport path from one emitter, wet deposition of NO_X-NO_3^- at the nearer square is calculated using the higher scavenging coefficient while the probability that NO_X-NO_3^- survives wet deposition to arrive at the farther square is calculated using the reduced coefficient. This causes some fraction of the emissions to be counted both as being wet deposited at the nearer square and as surviving to be wet deposited at the farther square. Thus, mass conservation for NO_X emissions and deposition is not imposed properly in this treatment.

The above description of NO_X-NO_3^- washout is parameterized with a scavenging coefficient meant to apply for NO_3^-, which is used to calculate wet deposition fluxes at the receptor. A dimensionless reduction factor multiplies the NO_3^- scavenging coefficient to give an effective scavenging coefficient which is applied to calculate the probability that a particle of NO_X-NO_3^- survives wet deposition along the transport path.

The parameter values were adjusted in a subjective way, with no rigorous least-squares fitting done to minimize the residual. The modelled quantities were found in general not to depend very sensitively upon the chosen values of parameters. Rather large changes were needed to obtain significantly changed results.

The parameter values obtained in this way are summarized in Table VI. The values of modelled wet deposition fluxes obtained with this parameter set are given in Table A.3 along with observed values. Ambient air concentrations are presented in Table A.4.

5.4 Comparison of Results to Observations

Excluding the Essex and Abitibi sites from consideration for the reasons mentioned in Section 3.0, the modelled values for wet deposition fluxes differ from the observed values by no more than 29% and typically by 20% for $SO_4^=$ and by no more than 36% and typically 25% for NO_3^-. Air concentrations differ from observed values by no more than 50% and typically by 25% for NO_3^- and by no more than 16% and typically 3% for $SO_4^=$. Modelled values both overpredict and underpredict the observed values, except for NO_3^- wet deposition, which is consistently underestimated. The apprehended difficulty with violation of mass conservation for NO_x-NO_3^- is in these cases not serious.

6.0 ALTERED CHEMICAL PARAMETERS: DISCUSSION

The degree of agreement between modelled and observed values is quite good. However, given the large differences between the NO_x-NO_3^- scavenging coefficient, the NO_x oxidation rate constant, and the SO_2 dry deposition velocity of the adjusted parameter set (Table VI) and the base parameter set (Table V), the model when using these parameters must be regarded as an empirical fit to observed values rather than as an accurate model of the physical processes involved.

Comparisons were done using the parameters from Table VI between modelled and observed values for the years 1978, 1979, 1980 and 1981, and are given in Tables A.5, A.6 and A.7. Agreement is not quite as good as for the year 1979 for which the fit was done, but is still acceptable.

7.0 ESTIMATES OF HYDRO's CONTRIBUTIONS TO ACID DEPOSITION IN SENSITIVE REGIONS

Tables A.8-A.15 summarize the results calculated by Fisher's statistical long-range transport model for all sources and Hydro sources only for the years 1978-1981 using the altered chemical parameter set. Estimated Hydro percentage contributions are presented in Table A.16. Results are presented for five sensitive receptor regions illustrated in Figure 4; Lakehead (grid squares (3,15), (3,16), (4,15), (4,16)), Upper State Michigan ((5,12), (5,13), (6,12), (6,13)), Algoma ((8,13), (8,14), (9,13), (9,14)), Muskoka-Haliburton ((12,11), (12,12), (13,11), (13,12), (14,11)) and the Adirondacks ((15,9), (15,10), (16,9), (16,10)).

TABLE VI

ALTERED CHEMICAL PARAMETERS

λ (Rainfall Scavenging Coefficient, s^{-1})*

SO_2	3×10^{-4}
$SO_4^=$	3×10^{-4}
NO_x	1×10^{-3}
NO_3^-	1×10^{-3}

Reduction factor for NO_x-NO_3^- scavenging in transport: 0.05

*For assumed rainfall intensity of 1 mm h^{-1}.

Dry Deposition Velocities (m s^{-1})

SO_2	0.001
$SO_4^=$	0.0
NO_x	0.0038
NO_3^-	0.04

Transformation Coefficients (s^{-1})

$SO_2 \rightarrow SO_4^=$	1.5×10^{-6}
$NO_x \rightarrow NO_3^-$	1.0×10^{-6}

Fraction Initially Converted

$SO_2 \rightarrow SO_4^=$	0.03
$NO_x \quad NO_3^-$	0.0

8.0 RECOMMENDATIONS

Before the results of the model can be trusted as giving an accurate estimate of Ontario Hydro's contribution to acid deposition fluxes, a better understanding must be gained of the ways in which the parameterization set adopted here compensates for physical or chemical processes ignored or poorly represented in the model. Even though total deposition fluxes in sensitive areas are well predicted, the relative contributions of near and far sources may not be accurately predicted.

For this reason we recommend further development of the model, including the following points:

(1) Inclusion of different effective release heights for S and N compounds.

(2) Use of the true Sudbury stack height for emissions there.

(3) Examine precipitation statistics to determine the effect of including seasonal and spatial variations in average precipitation intensity and average duration of wet and dry periods.

(4) Investigate Fisher's and other (eg, Venkatram et al, 1982) statistical methods of accounting for washout during transport, in order to find a means to incorporate different precipitation scavenging coefficients for interconvertible species.

(5) Carefully constructing gridded emissions inventories for some additional years to provide more tests of accuracy.

(6) Examine the possibility of using more than a single wind frequency distribution over the gridded area.

(7) Develop improved nitrogen chemistry.

9.0 CONCLUSIONS

The Fisher statistical long-range transport model as initially parameterized to include both S and N transport does not adequately predict North American wet deposition fluxes or ambient aerosol concentrations, with the former significantly underestimated and the latter significantly overestimated.

An altered parameterization provides good predictions of these quantities but the physical or chemical validity of this parameter set is questionable.

It is reasonably likely that further examination both of the representation of important processes in the Fisher model and of the values used for several meteorological and emissions parameters might yield a model giving predictions of acid deposition fluxes as accurate as those obtained in this work and yet employing physically sensible representations of the relevant processes.

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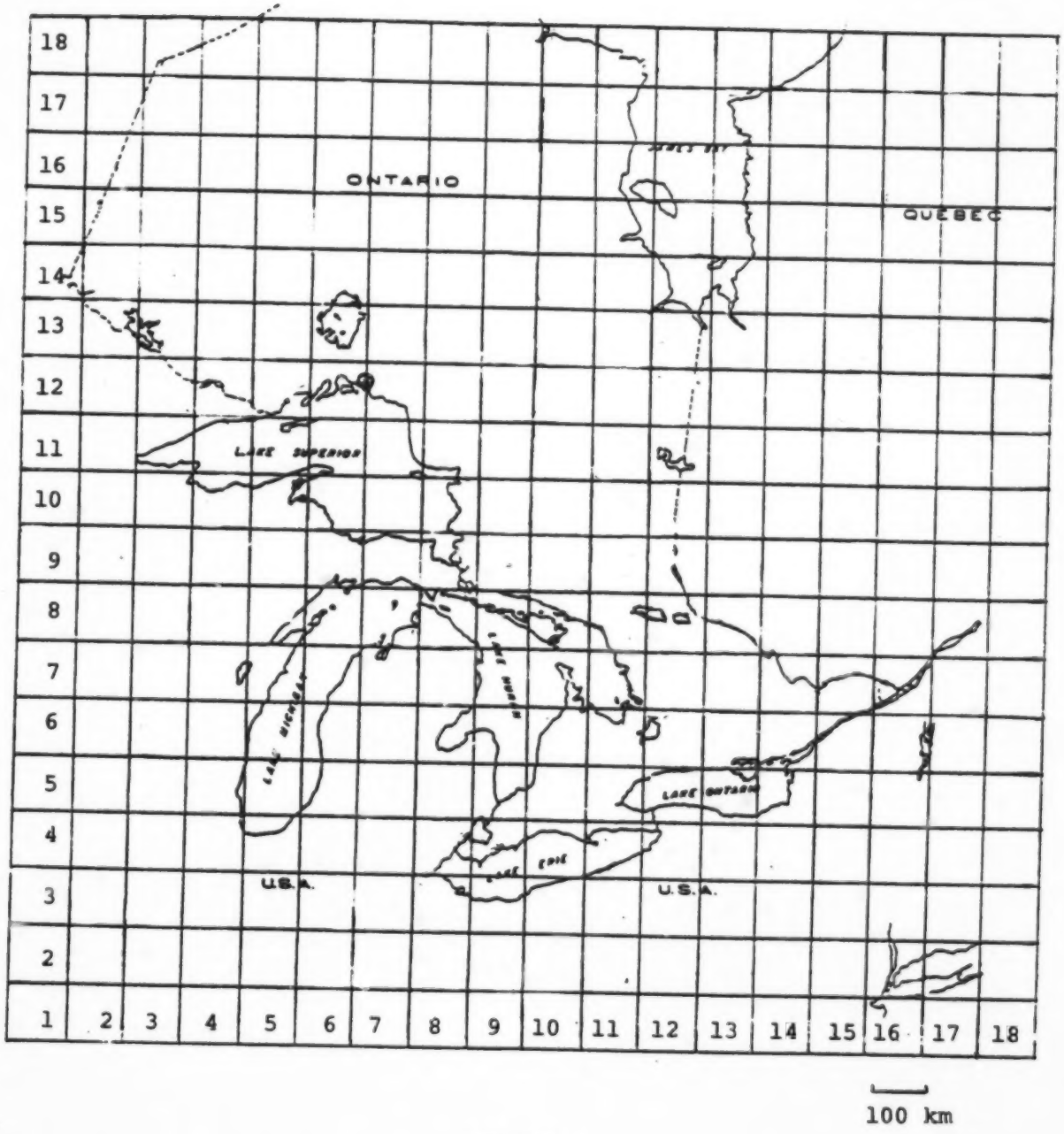

FIGURE 1

ORIGINAL EMISSIONS AREA

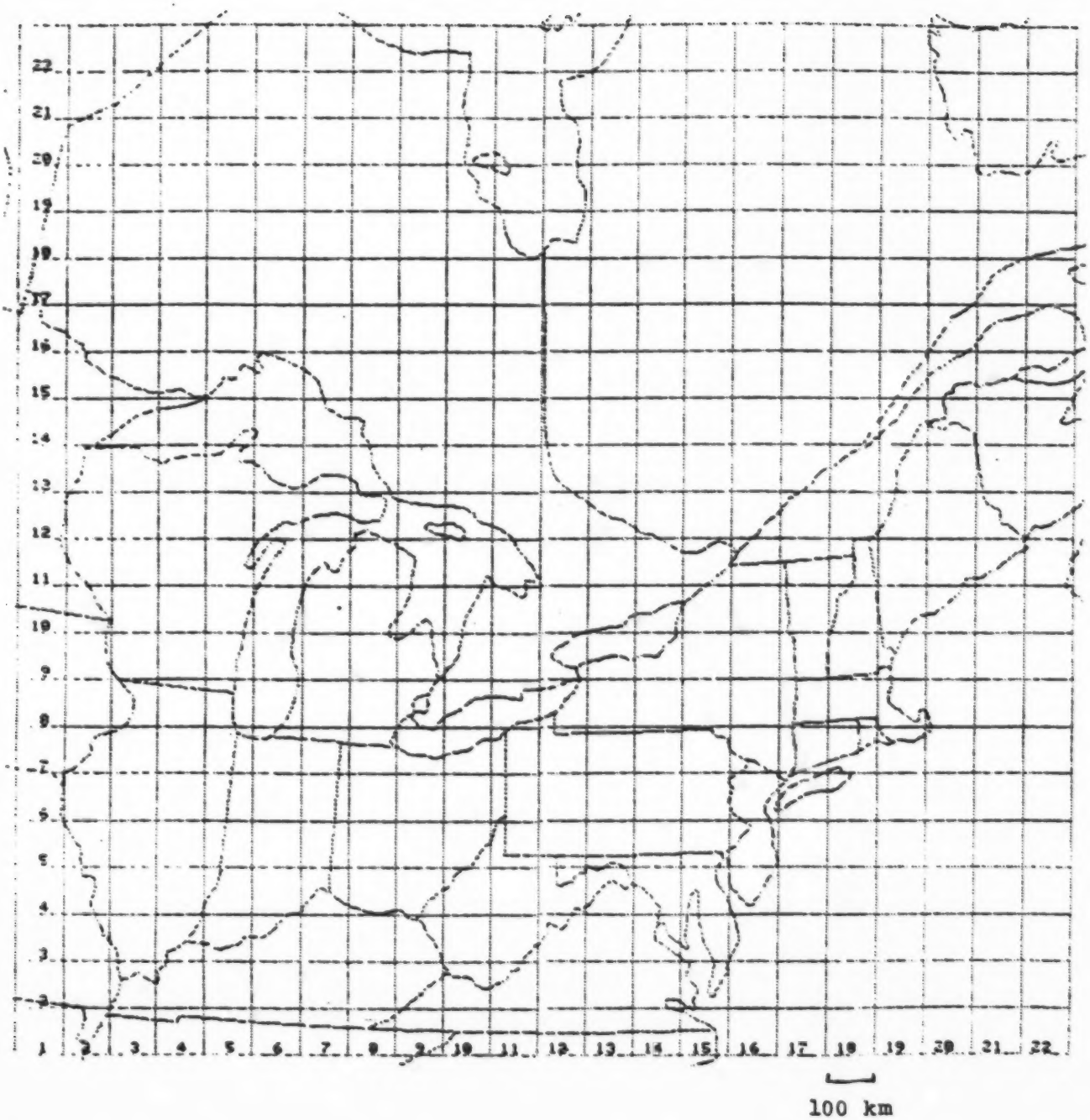

FIGURE 2

EXPANDED EMISSIONS AREA

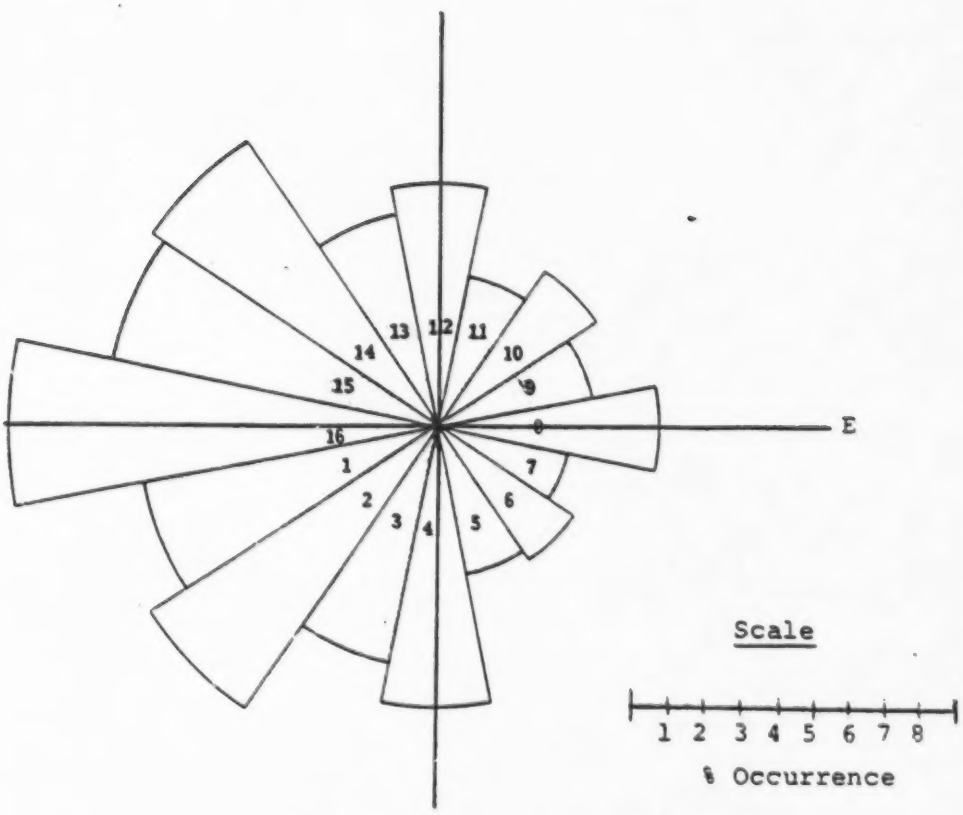

FIGURE 3
WINDROSE FOR STUDY AREA

154286-RD

Sensitive Receptor Areas:

1 Lakehead
2 Upper State Michigan
3 Algoma
4 Haliburton/Muskoka
5 Adirondacks

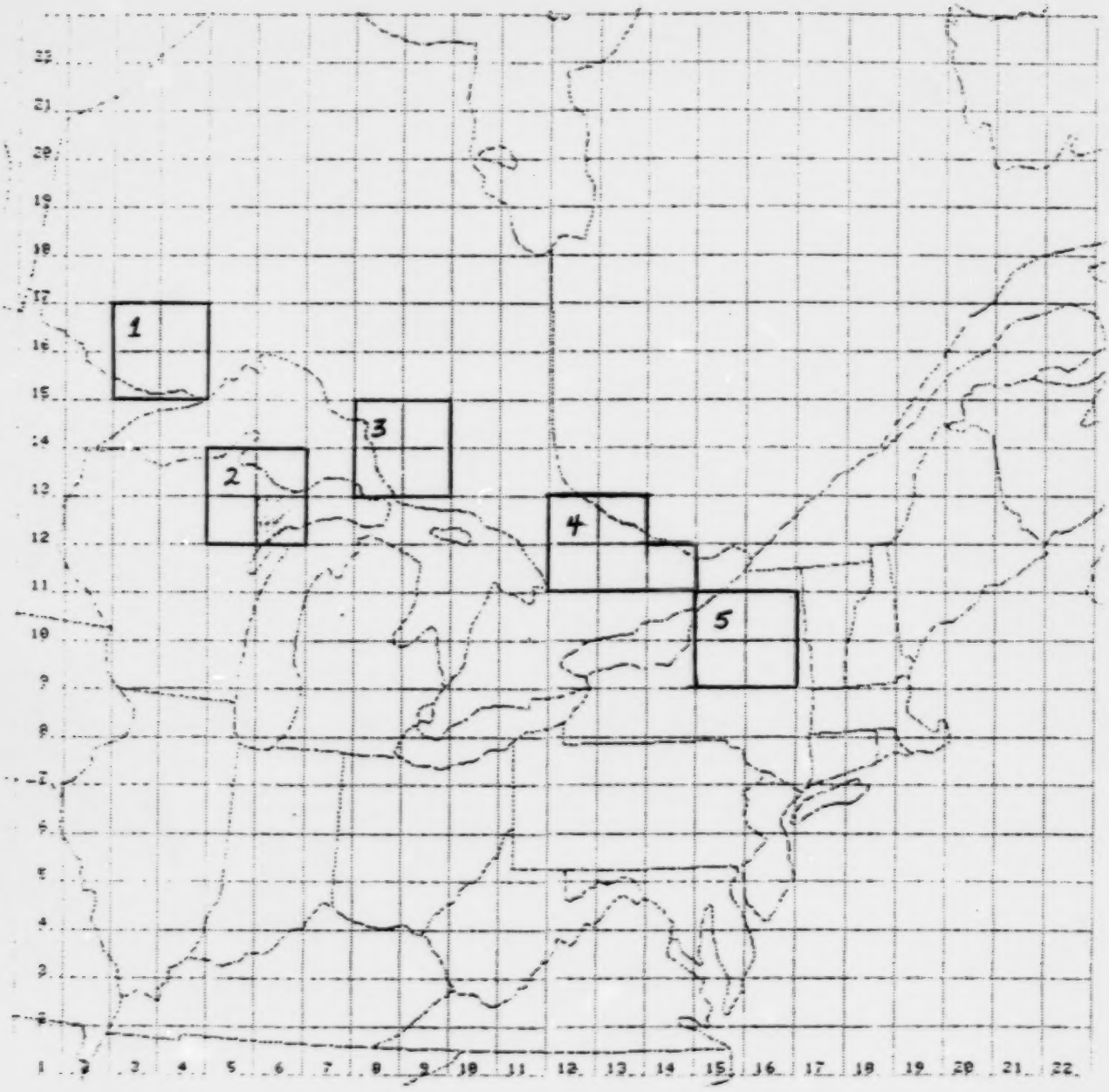

FIGURE 4

TABLE A.1

$SO_4^=$, NO_3^-, WET DEPOSITION, 1979 (g/m^2 YEAR)
(INITIALLY-CHOSEN MODEL PARAMETERS)

Grid Square x y	Measurement Location	1979 $SO_4^=$ (Wet)		1979 NO_3^- (Wet)	
		Modelled (No changes)	Measured	Modelled (No changes)	Measured
9, 12	Mississagi	2.19	4.7	1.22	2.7
10, 17	Abitibi	0.73	3.1	0.49	1.5
12, 8	Peacock Pt	5.22	7.2	2.43	4.9
12, 9	Toronto	4.32	3.9	2.20	3.0
	Burlington		5.0		4.1
12, 11	Muskoka	3.53	4.0	1.67	3.1
15, 11	Vankleek Hill	2.63	4.8	1.43	3.3

TABLE A.2

$SO_4^=$, NO_3^-, AIR CONCENTRATIONS, 1979 ($\mu g/m^3$)
(INITIALLY CHOSEN MODEL PARAMETERS)

(Annual Averages)

Grid Square x y	Measurement Location	1979 $SO_4^=$		1979 NO_3	
		Modelled (No changes)	Measured	Modelled (No changes)	Measured
9, 8	Essex	10.25	10.9	34.73	6.7
9, 12	Mississagi	7.10	5.4	20.28	1.5
10, 17	Abitibi	4.23	3.1	8.71	0.4
12, 8	Peacock Pt	11.27	8.0	37.67	3.6
12, 9	Toronto	10.08	7.4	33.35	4.5
	Burlington		7.5		4.1
	Markham		7.0		4.2
	Welland		8.4		4.3
12, 10	Fenelon Falls			29.95	2.3
12, 11	Muskoka	8.93	6.7	27.07	1.8
15, 11	Vankleek Hill	7.96	7.4	23.36	3.8

TABLE A.3

$SO_4^=$, NO_3^-, WET DEPOSITION, 1979 (g/m² year) (ALTERED MODEL PARAMETERS)

Grid Square x y	Measurement Location	1979 $SO_4^=$ (Wet)		1979 NO_3^- (Wet)	
		Modelled (Altered)	Measured	Modelled (Altered)	Measured
9, 12	Mississagi	3.54	4.7	2.09	2.7
10, 17	Abitibi	1.34	3.1	1.40	1.5
12, 8	Peacock Pt	7.63	7.2	3.23	4.9
12, 9	Toronto	6.4	3.9	2.96	3.0
	Burlington		5.0		4.1
12, 11	Muskoka	5.30	4.0	2.59	3.1
15, 11	Vankleek Hill	4.05	4.8	2.47	3.3

TABLE A.4

SULFATE AND NITRATE AEROSOL CONCENTRATIONS, 1979 (µg/m³) (ALTERED MODEL PARAMETERS)

(Annual Averages)

Grid Square x y	Measurement Location	1979 $SO_4^=$		1979 NO_3^-	
		Modelled (Altered)	Measured	Modelled (Altered)	Measured
9, 8	Essex	8.88	10.9	2.10	6.7
9, 12	Mississagi	5.54	5.4	1.63	1.47
10, 17	Abitibi	3.37	3.1	1.10	0.4
12, 8	Peacock Pt	8.96	8.0	2.44	3.6
12, 9	Toronto	7.98	7.4	2.21	4.5
	Burlington		7.5		4.1
	Markham		7.0		4.2
	Welland		8.4		3.3
12, 10	Fenelon Falls			2.07	2.3
12, 11	Muskoka	7.1	6.7	2.01	1.8
15, 11	Vankleek Hill	6.3	7.4	1.91	3.8

TABLE A.5

$SO_4^=$ AIR CONCENTRATIONS ($\mu g/m^3$) 1978-81
(ALTERED PARAMETERS)

(Annual Averages)

Grid Square x y	Measurement Location	1978		1979		1980		1981	
		Modelled	Measured	Modelled	Measured	Modelled	Measured	Modelled	Measured
9, 8	Essex	8.11	16.1	8.88	10.9	8.86	9.4	6.68	11.3
9, 12	Mississagi	-	-	5.54	5.4	5.52	3.9	4.52	1.6
10,16	Kapuskasing	-	-	-	-	-	-	3.02	3.4
10,17	Abitibi	-	-	3.37	3.1	-	-	-	-
12,8	Peacock Pt	9.5	7.7	8.96	8.0	8.89	5.9	-	-
12,9	Toronto	8.54	9.5	7.98	7.4	7.92	6.0	-	-
	Burlington		9.0		7.5		5.6		-
	Markham		7.0		7.0		4.1		-
	Welland		7.5		8.4		-		-
12,10	Fenelon Falls	7.23	7.0	-	-	-	-	-	-
12,11	Muskoka	-	-	7.1	6.7	7.06	4.3	5.79	4.2
15,11	Vankleek Hill	-	-	6.3	7.4	6.28	6.1	5.21	7.9

TABLE A.6

NO_3^- AIR CONCENTRATION ($\mu g/m^3$) 1978-81
(ALTERED PARAMETERS)

(Annual Averages)

Grid Square x y	Measurement Location	1978		1979		1980		1981	
		Modelled	Measured	Modelled	Measured	Modelled	Measured	Modelled	Measured
9, 8	Essex	1.90	10.3	2.10	6.7	2.10	5.61	1.80	2.8
9, 12	Mississagi	1.48	1.9	1.63	1.1	1.63	1.47	1.41	0.6
10,16	Kapuskasing	-	-	-	-	-	-	1.01	0.9
10,17	Abitibi	1.01	0.7	1.10	0.4	-	-	-	-
12, 8	Peacock Pt	2.12	2.4	2.44	3.6	2.44	2.49	-	-
12, 9	Toronto	1.97	4.8	2.21	4.5	2.22	2.93	-	-
	Burlington		3.9		4.1		3.99		-
	Markham		4.1		4.2		2.69		-
	Welland		3.1		3.3		-		-
	Cayuga		6.6		-		-		-
12,10	Fenelon Falls	1.64	2.7	2.07	2.3	-	-	-	-
12,11	Muskoka	-	-	2.01	1.8	2.01	1.33	1.67	0.7
13,10	Peterborough	1.81	2.6	-	-	-	-	-	-
	Frankford		2.9		-		-		-
15,11	Vankleek Hill	1.62	5.6	1.91	3.8	1.91	3.88	1.59	1.8

TABLE A.7

$SO_4^=$ AND NO_3^- WET DEPOSITION (g/m^2 year) 1978-79
(ALTERED PARAMETERS)

Grid Square x y	Measurement Location	$SO_4^=$				NO_3^-			
		1978		1979		1978		1979	
		Modelled	Measured	Modelled	Measured	Modelled	Measured	Modelled	Measured
9, 12	Mississagi	-	-	3.54	4.7	-	-	2.09	2.7
10,17	Abitibi	-	-	1.34	3.1	-	-	1.40	1.5
12, 8	Peacock Pt	9.15	5.0	7.63	7.2	2.79	4.2	3.23	4.9
12, 9	Toronto	7.29	3.3	6.40	3.9	2.79	2.3	2.96	3.0
	Burlington		4.6		5.0		3.7		4.1
12,10	Fenelon Falls	5.16	4.6	-	-	2.35	3.0	-	-
12,11	Muskoka	-	-	5.30	4.0	-	-	2.59	3.1
15,11	Vankleek Hill	-	-	4.05	4.8	-	-	2.47	3.3

TABLE A.8

MODELLED ANNUAL DEPOSITION AND AEROSOL CONCENTRATIONS - 1978, ALL SOURCES; ALTERED PARAMETERS

Map Square x	y	Wet dep $SO_2 + SO_4^=$ ($g(SO_4^=)/m^2$)	Wet dep NO_3^- ($g(NO_3^-)/m^2$)	H^+ equiv $SO_4^= + NO_3^-$ ($mequiv/m^2$)	$SO_4^=$ aerosol conc ($\mu g(SO_4^=)/m^3$)	NO_3^- aerosol conc ($\mu g(NO_3^-)/m^3$)
Region 1: Lakehead						
3	15	1.41	1.11	43.37	3.04	0.86
3	16	1.17	1.04	40.14	2.74	0.82
4	15	1.55	1.27	49.32	3.35	0.99
4	16	1.25	1.16	44.47	2.95	0.91
Mean:		1.35	1.14	44.33	3.02	0.90
Region 2: Upper Michigan						
5	12	2.99	1.64	67.17	4.60	1.26
5	13	2.20	1.47	58.39	4.09	1.14
6	12	2.73	1.69	67.68	4.73	1.31
6	13	2.36	1.64	64.79	4.42	1.28
Mean:		2.57	1.61	64.51	4.46	1.24
Region 3: Algoma						
8	13	2.63	1.67	65.94	4.68	1.30
8	14	2.19	1.60	62.36	4.36	1.26
9	13	3.10	1.74	69.28	5.04	1.36
9	14	2.60	1.62	63.75	4.68	1.26
Mean:		2.63	1.66	65.33	4.69	1.29
Region 4: Muskoka-Haliburton						
12	11	4.88	2.24	91.20	7.13	1.75
12	12	5.17	2.09	84.71	6.90	1.64
13	11	4.12	2.18	87.87	6.71	1.70
13	12	3.78	2.02	80.47	6.23	1.58
14	11	3.74	2.15	85.91	6.43	1.68
Mean:		4.34	2.14	86.03	6.68	1.67
Region 5: Adirondacks						
15	9	4.52	2.40	97.43	7.19	1.86
15	10	3.99	2.23	90.35	6.81	1.74
16	9	4.39	2.50	101.43	7.05	1.93
16	10	3.73	2.24	90.17	6.45	1.75
Mean:		4.16	2.34	94.97	6.87	1.82

TABLE A.9

MODELLED ANNUAL DEPOSITION AND AEROSOL CONCENTRATIONS - 1979, ALL SOURCES; ALTERED PARAMETERS

Map Square x	y	Wet dep $SO_2 + SO_4^=$ ($g(SO_4^=)/m^2$)	Wet dep NO_3^- ($g(NO_3^-)/m^2$)	H^+ equiv $SO_4^= + NO_3^-$ ($mequiv/m^2$)	$SO_4^=$ aerosol conc ($\mu g(SO_4^=)/m^3$)	NO_3^- aerosol conc ($\mu g(NO_3^-)/m^3$)
Region 1:	Lakehead					
3	15	1.33	1.13	43.54	2.74	0.88
3	16	1.09	1.07	40.66	2.46	0.84
4	15	1.49	1.29	49.22	3.04	1.01
4	16	1.18	1.18	44.38	2.67	0.92
Mean:		1.27	1.17	44.45	2.72	0.91
Region 2:	Upper Michigan					
5	12	3.14	1.66	67.03	4.39	1.28
5	13	2.16	1.49	58.50	3.79	1.16
6	12	2.75	1.72	68.19	4.48	1.34
6	13	2.31	1.65	64.35	4.12	1.29
Mean:		2.59	1.63	64.52	4.20	1.27
Region 3:	Algoma					
8	13	2.39	1.78	68.93	4.43	1.39
8	14	2.25	1.29	68.81	4.44	1.40
9	13	2.77	1.93	75.11	4.90	1.51
9	14	2.42	1.77	69.01	4.58	1.40
Mean:		2.46	1.82	70.46	4.59	1.42
Region 4:	Muskoka-Haliburton					
12	11	5.30	2.59	105.59	7.10	2.01
12	12	6.07	2.42	97.68	7.08	1.89
13	11	4.62	2.60	104.61	6.95	2.02
13	12	4.24	2.33	92.59	6.39	1.82
14	11	4.41	2.57	102.64	6.58	1.99
Mean:		4.93	2.50	100.62	6.82	1.95
Region 5:	Adirondacks					
15	9	4.91	2.90	117.64	7.24	2.24
15	10	4.86	2.62	105.95	6.92	2.03
16	9	4.65	3.03	121.71	7.06	2.32
16	10	4.13	2.66	106.65	6.46	2.06
Mean:		4.64	2.80	112.99	6.92	2.16

TABLE A.10

MODELLED ANNUAL DEPOSITION AND AEROSOL CONCENTRATIONS - 1980, ALL SOURCES; ALTERED PARAMETERS

Map Square x	Map Square y	Wet dep $SO_2 + SO_4^=$ ($g(SO_4^=)/m^2$)	Wet dep NO_3^- ($g(NO_3^-)/m^2$)	H^+ equiv $SO_4^= + NO_3^-$ ($mequiv/m^2$)	$SO_4^=$ aerosol conc ($\mu g(SO_4^=)/m^3$)	NO_3^- aerosol conc ($\mu g(NO_3^-)/m^3$)
Region 1:	Lakehead					
3	15	1.33	1.13	43.58	2.72	0.88
3	16	1.09	1.07	40.70	2.45	0.84
4	15	1.49	1.29	49.27	3.03	1.01
4	16	1.18	1.18	44.42	2.66	0.93
Mean:		1.27	1.17	44.49	2.71	0.91
Region 2:	Upper Michigan					
5	12	3.14	1.66	67.04	4.38	1.29
5	13	2.16	1.49	58.55	3.78	1.17
6	12	2.75	1.72	68.25	4.47	1.34
6	13	2.31	1.65	64.40	4.11	1.29
Mean:		2.59	1.63	64.56	4.18	1.27
Region 3:	Algoma					
8	13	2.39	1.78	69.00	4.42	1.39
8	14	2.25	1.79	68.87	4.42	1.40
9	13	2.77	1.93	75.18	4.89	1.51
9	14	2.41	1.79	69.07	4.56	1.40
Mean:		2.45	1.82	70.53	4.57	1.42
Region 4:	Muskoka-Haliburton					
12	11	5.27	2.60	105.75	7.06	2.01
12	12	6.04	2.42	97.77	7.05	1.89
13	11	4.59	2.60	104.69	6.92	2.02
13	12	4.22	2.34	92.67	6.36	1.82
14	11	4.37	2.57	102.87	6.54	2.00
Mean:		4.90	2.51	100.75	6.79	1.95
Region 5:	Adirondacks					
15	9	4.87	2.91	117.65	7.19	2.25
15	10	4.82	2.63	106.05	6.87	2.03
16	9	4.62	3.03	121.70	7.01	2.33
16	10	4.10	2.67	106.62	6.42	2.06
Mean:		4.60	2.81	113.00	6.87	2.17

TABLE A.11

MODELLED ANNUAL DEPOSITION AND AEROSOL CONCENTRATIONS - 1981, ALL SOURCES; ALTERED PARAMETERS

Map Square x	Map Square y	Wet dep $SO_2 + SO_4^=$ ($g(SO_4^=)/m^2$)	Wet dep NO_3^- ($g(NO_3^-)/m^2$)	H^+ equiv $SO_4^= + NO_3^-$ ($mequiv/m^2$)	$SO_4^=$ aerosol conc ($\mu g(SO_4^=)/m^3$)	NO_3^- aerosol conc ($\mu g(NO_3^-)/m^3$)
Region 1: Lakehead						
3	15	1.06	1.03	39.09	2.27	0.80
3	16	0.87	0.97	36.37	2.04	0.76
4	15	1.21	1.18	44.60	2.55	0.92
4	16	0.96	1.08	40.34	2.25	0.85
Mean:		1.02	1.06	40.10	2.28	0.83
Region 2: Upper Michigan						
5	12	2.45	1.55	61.49	3.63	1.20
5	13	1.75	1.41	54.24	3.18	1.10
6	12	2.19	1.56	61.09	3.70	1.22
6	13	1.85	1.51	57.89	3.41	1.18
Mean:		2.06	1.51	58.68	3.48	1.17
Region 3: Algoma						
8	13	1.92	1.56	59.63	3.60	1.22
8	14	1.77	1.53	58.04	3.52	1.20
9	13	2.19	1.63	62.58	3.91	1.27
9	14	1.95	1.53	58.19	3.68	1.19
Mean:		1.96	1.56	59.61	3.68	1.22
Region 4: Muskoka-Haliburton						
12	11	4.44	2.17	87.95	5.79	1.67
12	12	5.38	2.03	81.77	5.91	1.58
13	11	3.84	2.16	86.93	5.64	1.68
13	12	3.62	1.95	77.02	5.25	1.52
14	11	3.75	2.15	86.03	5.40	1.67
Mean:		4.21	2.09	83.94	5.60	1.62
Region 5: Adirondacks						
15	9	4.05	2.40	97.55	5.82	1.86
15	10	4.18	2.21	89.54	5.72	1.71
16	9	3.81	2.49	100.10	5.69	1.91
16	10	3.49	2.19	88.01	5.30	1.69
Mean:		3.88	2.32	93.80	5.63	1.79

TABLE A.12

MODELLED ANNUAL DEPOSITION AND AEROSOL CONCENTRATIONS - 1978, HYDRO SOURCES; ALTERED PARAMETERS

Map Square x	Map Square y	Wet dep $SO_2 + SO_4^=$ ($g(SO_4^=)/m^2$)	Wet dep NO_3^- ($g(NO_3^-)/m^2$)	H^+ equiv $SO_4^= + NO_3^-$ ($mequiv/m^2$)	$SO_4^=$ aerosol conc ($\mu g(SO_4^=)/m^3$)	NO_3^- aerosol conc ($\mu g(NO_3^-)/m^3$)
Region 1:	**Lakehead**					
3	15	0.020	0.007	0.311	0.045	0.005
3	16	0.017	0.006	0.297	0.042	0.005
4	15	0.029	0.008	0.363	0.052	0.006
4	16	0.021	0.007	0.326	0.046	0.005
Mean:		0.022	0.007	0.324	0.046	0.005
Region 2:	**Upper Michigan**					
5	12	0.031	0.008	0.375	0.055	0.006
5	13	0.034	0.009	0.416	0.061	0.007
6	12	0.044	0.010	0.478	0.071	0.008
6	13	0.039	0.009	0.449	0.066	0.007
Mean:		0.037	0.009	0.430	0.063	0.007
Region 3:	**Algoma**					
8	13	0.052	0.010	0.524	0.077	0.008
8	14	0.042	0.011	0.476	0.070	0.008
9	13	0.059	0.011	0.555	0.082	0.009
9	14	0.045	0.011	0.490	0.072	0.008
Mean:		0.049	0.011	0.511	0.075	0.008
Region 4:	**Muskoka-Haliburton**					
12	11	0.235	0.024	1.393	0.215	0.018
12	12	0.143	0.020	1.081	0.160	0.016
13	11	0.183	0.021	1.194	0.182	0.016
13	12	0.136	0.020	1.069	0.163	0.016
14	11	0.205	0.027	1.440	0.213	0.020
Mean:		0.180	0.022	1.235	0.186	0.016
Region 5:	**Adirondacks**					
15	9	0.206	0.028	1.528	0.229	0.022
15	10	0.891	0.024	1.291	0.195	0.018
16	9	0.142	0.026	1.309	0.194	0.020
16	10	0.132	0.023	1.178	0.179	0.018
Mean:		0.168	0.025	1.326	0.199	0.019

TABLE A.13

MODELLED ANNUAL DEPOSITION AND AEROSOL CONCENTRATIONS - 1979, HYDRO SOURCES; ALTERED PARAMETERS

Map Square x	y	Wet dep $SO_2 + SO_4^=$ ($g(SO_4^=)/m^2$)	Wet dep NO_3^- ($g(NO_3^-)/m^2$)	H^+ equiv $SO_4^= + NO_3^-$ ($mequiv/m^2$)	$SO_4^=$ aerosol conc ($\mu g(SO_4^=)/m^3$)	NO_3^- aerosol conc ($\mu g(NO_3^-)/m^3$)
Region 1:	Lakehead					
3	15	0.022	0.007	0.333	0.047	0.006
3	16	0.018	0.007	0.314	0.043	0.005
4	15	0.034	0.008	0.395	0.055	0.006
4	16	0.024	0.007	0.348	0.048	0.006
Mean:		0.025	0.007	0.347	0.048	0.006
Region 2:	Upper Michigan					
5	12	0.033	0.008	0.398	0.057	0.006
5	13	0.037	0.009	0.443	0.064	0.007
6	12	0.047	0.010	0.503	0.073	0.008
6	13	0.042	0.010	0.474	0.069	0.008
Mean:		0.040	.009	0.454	0.066	0.007
Region 3:	Algoma					
8	13	0.055	0.011	0.556	0.080	0.009
8	14	0.045	0.010	0.507	0.072	0.008
9	13	0.061	0.012	0.585	0.085	0.009
9	14	0.047	0.010	0.516	0.074	0.008
Mean:		0.052	0.011	0.541	0.078	0.009
Region 4:	Muskoka-Haliburton					
12	11	0.246	0.026	1.466	0.224	0.019
12	12	0.146	0.021	1.123	0.163	0.016
13	11	0.183	0.022	1.235	0.184	0.017
13	12	0.139	0.021	1.117	0.168	0.016
14	11	0.187	0.028	1.446	0.207	0.021
Mean:		0.180	0.024	1.277	0.189	0.018
Region 5:	Adirondacks					
15	9	0.196	0.030	1.561	0.228	0.023
15	10	0.162	0.024	1.265	0.183	0.019
16	9	0.138	0.027	1.344	0.194	0.021
16	10	0.122	0.023	1.190	0.175	0.018
Mean:		0.154	0.026	1.340	0.195	1.020

TABLE A.14

MODELLED ANNUAL DEPOSITION AND AEROSOL CONCENTRATIONS - 1980, HYDRO SOURCES; ALTERED PARAMETERS

Map Square x	Map Square y	Wet dep $SO_2 + SO_4^=$ ($g(SO_4^=)/m^2$)	Wet dep NO_3^- ($g(NO_3^-)/m^2$)	H^+ equiv $SO_4^= + NO_3^-$ ($mequiv/m^2$)	$SO_4^=$ aerosol conc ($\mu g(SO_4^=)/m^3$)	NO_3^- aerosol conc ($\mu g(NO_3^-)/m^3$)
Region 1:	Lakehead					
3	15	0.021	0.011	0.461	0.043	0.009
3	16	0.017	0.011	0.438	0.040	0.009
4	15	0.032	0.013	0.536	0.051	0.010
4	16	0.022	0.012	0.481	0.044	0.009
Mean:		0.023	0.012	0.479	0.045	0.009
Region 2:	Upper Michigan					
5	12	0.031	0.013	0.549	0.052	0.010
5	13	0.034	0.015	0.614	0.058	0.011
6	12	0.043	0.016	0.703	0.067	0.013
6	13	0.039	0.016	0.660	0.063	0.012
Mean:		0.037	0.015	0.631	0.060	0.012
Region 3:	Algoma					
8	13	0.051	0.018	0.763	0.073	0.014
8	14	0.042	0.016	0.695	0.066	0.013
9	13	0.057	0.018	0.805	0.078	0.014
9	14	0.043	0.017	0.708	0.068	0.013
Mean:		0.048	0.017	0.743	0.071	0.014
Region 4:	Muskoka-Haliburton					
12	11	0.224	0.041	1.997	0.205	0.031
12	12	0.132	0.034	1.550	0.049	0.026
13	11	0.162	0.036	1.693	0.167	0.027
13	12	0.125	0.034	1.532	0.154	0.026
14	11	0.156	0.044	1.987	0.183	0.034
Mean:		0.160	0.038	1.752	0.172	0.029
Region 5:	Adirondacks					
15	9	0.168	0.046	2.100	0.203	0.036
15	10	0.124	0.038	1.696	0.157	0.029
16	9	0.120	0.042	1.823	0.174	0.032
16	10	0.102	0.037	1.609	0.154	0.029
Mean:		0.129	0.041	1.807	0.172	0.032

TABLE A.15

MODELLED ANNUAL DEPOSITION AND AEROSOL CONCENTRATIONS - 1981, HYDRO SOURCES; ALTERED PARAMETERS

Map Square x	Map Square y	Wet dep $SO_2 + SO_4^=$ ($g(SO_4^=)/m^2$)	Wet dep NO_3^- ($g(NO_3^-)/m^2$)	H^+ equiv $SO_4^= + NO_3^-$ ($mequiv/m^2$)	$SO_4^=$ aerosol conc ($\mu g(SO_4^=)/m^3$)	NO_3^- aerosol conc ($\mu g(NO_3^-)/m^3$)
Region 1:	Lakehead					
3	15	0.021	0.008	0.371	0.045	0.007
3	16	0.017	0.008	0.343	0.042	0.006
4	15	0.032	0.009	0.432	0.052	0.007
4	16	0.023	0.009	0.380	0.046	0.007
Mean:		0.023	0.009	0.382	0.046	0.007
Region 2:	Upper Michigan					
5	12	0.032	0.009	0.436	0.055	0.007
5	13	0.036	0.011	0.493	0.062	0.008
6	12	0.046	0.012	0.560	0.072	0.009
6	13	0.040	0.011	0.526	0.067	0.009
Mean:		0.039	0.011	0.504	0.06[illegible]	0.00[illegible]
Region 3:	Algoma					
8	13	0.052	0.013	0.599	0.076	0.011
8	14	0.043	0.012	0.550	0.069	0.009
9	13	0.059	0.013	0.632	0.081	0.010
9	14	0.045	0.012	0.560	0.071	0.009
Mean:		0.050	0.013	0.585	0.074	0.010
Region 4:	Muskoka-Haliburton					
12	11	0.224	0.028	1.521	0.210	0.021
12	12	0.133	0.024	1.179	0.153	0.018
13	11	0.164	0.025	1.296	0.173	0.019
13	12	0.128	0.020	1.181	0.159	0.018
14	11	0.158	0.031	1.503	0.189	0.024
Mean:		0.161	0.026	1.337	0.177	0.020
Region 5:	Adirondacks					
15	9	0.167	0.031	1.571	0.206	0.024
15	10	0.120	0.027	1.291	0.163	0.021
16	9	0.120	0.028	1.365	0.177	0.022
16	10	0.105	0.026	1.232	0.161	0.020
Mean:		0.130	0.028	1.365	0.177	0.022

TABLE A.16

HYDRO PERCENTAGE CONTRIBUTIONS: 1978-81; ALTERED PARAMETERS

Region	Wet SO_2 + SO_4				Wet NO_3^-				H^+ equiv				$SO_4^=$ conc				NO_3^- conc			
	78	79	80	81	78	79	80	81	78	79	80	81	78	79	80	81	78	79	80	81
1. Lakehead	1.6	2.0	1.8	2.3	0.60	0.59	1.03	0.85	0.70	0.78	1.1	0.95	1.5	1.8	1.7	2.0	0.55	0.66	0.98	0.84
2. Upper Michigan	1.4	1.5	1.4	1.89	0.60	0.55	0.92	0.73	0.66	0.70	1.0	0.86	1.4	1.6	1.4	1.84	0.56	0.55	0.94	0.68
3. Algoma	1.9	2.1	2.0	2.55	0.66	0.60	0.9	0.83	0.78	0.77	1.1	0.98	1.6	1.7	1.6	2.0	0.62	0.63	0.98	0.82
4. Muskoka-Haliburton	4.1	3.7	3.3	3.8	1.0	0.96	1.5	1.24	1.44	1.27	1.7	1.6	2.8	2.8	2.5	3.16	0.95	0.92	1.49	1.23
5. Adirondacks	4.0	3.3	2.8	3.35	1.06	0.92	1.5	1.2	1.4	1.18	1.6	1.45	2.9	2.8	2.5	3.14	1.04	0.93	1.47	1.23